



ELSEVIER

Nuclear Instruments and Methods in Physics Research A 437 (1999) 374–380

**NUCLEAR
INSTRUMENTS
& METHODS
IN PHYSICS
RESEARCH**
Section A

www.elsevier.nl/locate/nima

Geometry and surface treatment dependence of the light collection from LSO crystals[☆]

J.S. Huber^{a,*}, W.W. Moses^a, M.S. Andreaco^b, M. Loepe^b, C.L. Melcher^b, R. Nutt^b

^aLawrence Berkeley National Laboratory, University of California, Mail Stop 55-121, 1 Cyclotron Road, Berkeley, CA 94720, USA

^bCTI, Inc., Knoxville, TN 37932, USA

Received 23 August 1998; received in revised form 24 June 1999; accepted 30 June 1999

Abstract

We study the relative light collection efficiency for narrow LSO crystals as a function of surface finish and geometry. We explore both a specular and diffuse surface reflector finish, using LSO crystals that are either polished or etched. The crystals have a square cross-section with widths of 1.8, 2.2, 2.5, or 2.6 mm and lengths of 10, 20 or 30 mm. When optically coupled to a PMT on a square end and wrapped with Teflon on 5 sides, we excite them at 5 mm incremental depths with 511 keV photons and measure the photopeak position. The light collection is characterized by a maximum output (based on the photopeak position when excited at the PMT end) and a depth-dependent loss factor. Both the maximum output and loss factor are effectively independent of width, and the maximum output is only weakly dependent on length (6696 ± 612 , 5796 ± 432 , and 5328 ± 288 photons for 10, 20, and 30 mm lengths). The loss factor is effectively independent of length, with a 0, 18, 27, and 22% reduction at excitation depths of 0, 10, 20, and 30 mm. The light collection is only weakly dependent on surface finish if it is polished or etched beyond a minimum time. © 1999 Published by Elsevier Science B.V. All rights reserved.

1. Introduction

We investigate the light collection from narrow lutetium oxyorthosilicate (LSO) [1] crystals as

a function of geometry and surface finish in order to develop a high-resolution positron emission tomograph [2]. In particular, we study the depth dependence of light collection efficiency and pulse height resolution. The LSO crystals are either polished or etched with pyrophosphoric acid.

An estimate of light collection can be made using first principles, but this is only valid for polished crystals [3,4] as it assumes specular reflection with 100% efficiency. It is difficult to predict collection from first principles for crystals with a diffuse surface reflector finish, since the exact reflection form (usually assumed to be Lambertian) is not known and multiple bounces (typically 12) [5,6] amplify

[☆]This work was supported in part by the U.S. Department of Energy under Contract No. DE-AC03-76SF00098, in part by Public Health Service Grant Nos. P01-HL25840 and R01-CA67911, and in part by Breast Cancer Research Program of the University of California Grant No. 1RB-0068.

*Corresponding author. Tel.: +1-510-486-6445; fax: +1-510-486-4768.

E-mail address: jshuber@lbl.gov (J.S. Huber)

any discrepancies. Light collection in other scintillator materials has been investigated with the aid of Monte Carlo simulations and direct measurements [7–11]. However, the differences in these scintillators' index of refraction, emission wavelength, and material surface properties make a direct comparison with LSO impossible. Hence, we measure the light collection from LSO crystals with a range of geometries.

The light collection is also significantly influenced by both the type of external reflector (specular or diffuse) and the reflector coupling method (coupling over an air gap or by direct contact), but that study is beyond the scope of this paper. The results presented in this paper use several layers of Teflon tape wrapped over an air gap as the reflector for all crystals. In addition, we do not measure the absolute total light output of the scintillator, but confine the study to relative light collection efficiency and pulse height resolution as a function of geometry and surface finish.

2. Experimental method

The LSO crystals are wrapped with a few layers (~ 0.30 mm) of Teflon tape, then heated ($\sim 75^\circ\text{C}$) in the dark for at least 15 min and subsequently kept in the dark in order to minimize phosphorescence that would affect the photomultiplier tube gain. After returning to room temperature, they are optically coupled to a photomultiplier tube (PMT) on a small square end of 1.8, 2.2, 2.5, or 2.6 mm width. The PMT is masked with black tape to exclude light that penetrates the reflector and impinges on the photocathode. All crystals of each size are measured using a Hamamatsu R-2497 PMT with a high voltage of 1500 V, and all 30 mm long crystals are also measured with a Burle S83062F PMT at 1500 V to check for systematic errors associated with the PMT. These moderate voltage levels are used to provide good linearity. We excite the crystals with 511 keV photons, measuring the photopeak position and width (Fig. 1) with a spectroscopy ADC after amplifying the signal with a $1\ \mu\text{s}$ shaping time constant. The PMT is attached to a sliding stand whose position is set using 5 mm wide spacers, thus allowing us to

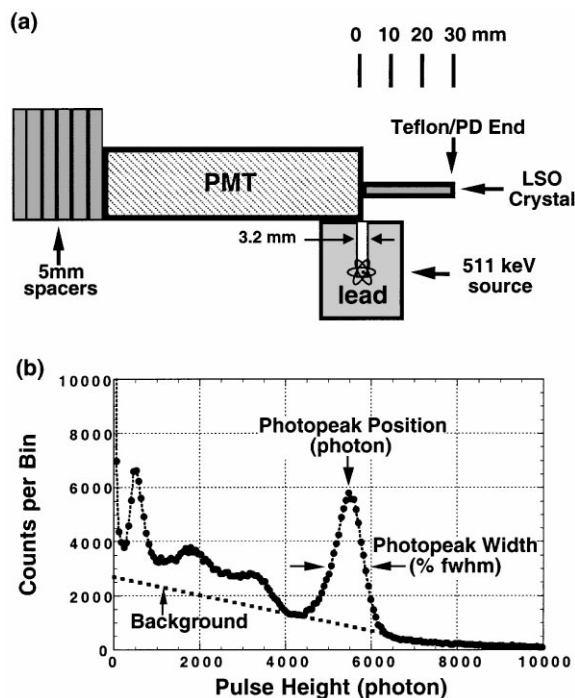


Fig. 1. (a) Experimental set-up. The LSO crystal is optically coupled to a photomultiplier tube and excited at 5 mm incremental depths with 511 keV photons. (b) A 511 keV pulse-height spectra of a typical LSO crystal as observed by a photomultiplier tube (solid line) and the corresponding background (dashed line) assumed to be linear.

excite the crystals at 5 mm incremental depths. The lead collimator consists of parallel plates with a $36.2\text{ mm} \times 3.2\text{ mm}$ slit. Since a LSO crystal typically couples onto the PMT at a distance of 1 cm from the collimator, an approximately 4 mm fwhm section of crystal is illuminated at each excitation depth.

The crystals are tested with Teflon wrapped on 5 sides to measure the “maximum output,” depth-dependent light loss factor, and pulse height resolution. The “maximum output” is defined as the center of the photopeak when the crystal is excited at zero depth (PMT end). The loss factor is the reduction in the photopeak position relative to the maximum output when excited at a certain depth. These relative light collection characteristics are measured as a function of etch time, crystal width, crystal length, and surface finish.

In order to minimize the systematic errors due to sample to sample variations and focus on surface treatment and geometry effects, we use multiple crystals (2–4) of varying widths (1.8, 2.2, and 2.6 mm) cut from adjacent pieces of the same boule of LSO. These are also compared with 2.5 mm wide crystals from a different boule of LSO. The reported results for each crystal size are thus an average over several individual crystals. In addition, each crystal is measured multiple times with the crystal being cleaned and re-wrapped with Teflon reflector between most measurements. We observe a typical variation of 10% between multiple measurements of the same crystal or different crystals with the same geometry and finish. Crystals of 30 mm length are etched or polished and tested, then cut into 2 pieces of 10 and 20 mm lengths for further tests.

The photopeak position is initially measured in terms of PMT ADC bins. To calibrate these measurements in terms of scintillation photons exiting the crystal, an LSO crystal is coupled to a photodiode to measure the photopeak position (in units of PD ADC bins). Direct interactions in the silicon photodiode from the 5.9 keV X-rays of an Fe-55 source are used to calibrate this amplifier output in units of electrons. The photodiode signal is then corrected for the photodiode quantum efficiency of 84% (which represents a weighted average over the LSO emission spectrum), giving an absolute photodiode signal level in terms of number of impinging photons. The photopeak position of the crystal is then measured with the PMT set-up when excited at 0 mm (Fig. 1). These calibration measurements are made for 4 different crystals of various widths (2.2–2.6 mm) and averaged. We convert all photopeak measurements to report in units of impinging photons.

3. Results

3.1. Etch time

The PET detector that we are developing will use over 20 000 LSO crystals. As a result, we prefer a chemically etched surface finish to a mechanical polish in order to reduce processing and handling

costs; chemical etches have been similarly developed for BGO [12] and GSO [13] scintillators. We etch the crystals in a 200°C pyrophosphoric acid bath, followed by a cleaning process consisting of a 5 min bath in boiling water, a 5 min bath in concentrated HCl, a water rinse, and an ethanol cleaning. The pyrophosphoric acid is prepared by heating concentrated phosphoric acid (85% in water) in an uncovered vessel until the water is driven off and its volume is reduced by $\geq 15\%$. We successively etch and test two crystals (widths of 2.2 and 2.6 mm) with a total etch time of 0, 0.5, 1, 2, 3, 5, 10, 12.5, and 15 min. Fig. 2 shows light collection curves for a typical crystal. The maximum output (i.e. light collection at 0 mm) is roughly independent of etch time with an average of 5328 ± 648 photons for all etch times, as shown in Fig. 2a.

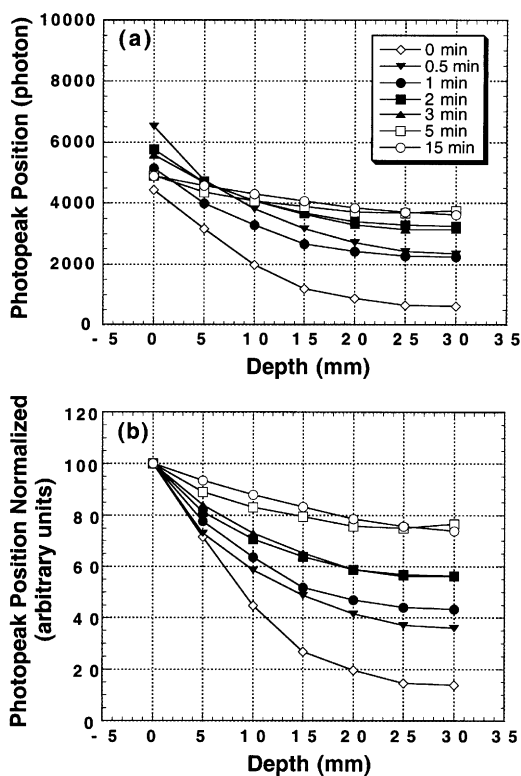


Fig. 2. (a) The photopeak position as a function of depth (PMT end at 0 mm) for a typical LSO crystal with Teflon wrapped on 5 sides. The crystal is successively etched for a total of 0, 0.5, 1, 2, 3, 5, 10, 12, and 15 min. (b) The same data as (a) except each curve is normalized to the same value at 0 mm depth.

Fig. 2b shows the photopeak position normalized into arbitrary units with a maximum output of 100, in order to eliminate the typical variations of 10% between multiple “identical” measurements. This figure demonstrates that the loss factor (i.e. the decrease in light collection as you excite closer to the 30 mm end) depends strongly on etch time. A crystal “as cut” has an 86% loss factor at 30 mm, which decreases to 24% for a crystal with a 5 min etch. Therefore, the etching can significantly improve the light collection homogeneity. The loss factor remains unchanged for etch times beyond 5 min; further etching reduces the crystal size without significantly effecting the surface finish. A 5 min etch is chosen for all “etched” crystals that we discuss here.

3.2. Crystal geometry

We test 2–4 etched crystals of each size and observe little crystal to crystal variation, thus we average the results for each crystal geometry. Table 1 lists this average photopeak position as a function of depth for etched crystals of 4 different widths (1.8, 2.2, 2.5, and 2.6 mm) and 3 different lengths (10, 20, and 30 mm) when Teflon is wrapped on all 5 sides. We observe only a small change in photopeak position as a function of depth. Both the maximum output and loss factor are effectively independent of the crystal width; this is particularly evident when comparing crystals from the same LSO boule with 1.8, 2.2, and 2.6 mm width. Therefore, we average the results over crystals of different

Table 1

The 511 keV photopeak position and energy resolution as a function of excitation depth (PMT end at 0 mm) for etched crystals of 4 different widths (1.8, 2.2, 2.5, and 2.6 mm) and 3 different lengths (10, 20, 30 mm) with Teflon wrapped on 5 sides. The photopeak position is in units of impinging photons, and photopeak resolution (shown in parentheses) is in units of fwhm %

Crystal dimensions (mm)		Excitation depth (mm)						
Width	Length	0	5	10	15	20	25	30
2.6	10	5688 (13)	5184 (14)	4824 (13)				
2.6	20	5436 (17)	4752 (20)	4104 (19)	3636 (15)	3528 (13)		
2.6	30	4860 (11)	4644 (14)	4320 (11)	4086 (8)	3816 (10)	3708 (12)	3708 (12)
2.5	10	7200 (15)	6660 (13)	6372 (13)				
2.5	20	5652 (14)	5148 (15)	4788 (14)	4536 (14)	4392 (12)		
2.5	30	5598 (11)	5292 (12)	5094 (12)	4932 (10)	4824 (10)	4734 (11)	4698 (11)
2.2	10	7128 (10)	6552 (14)	6336 (12)				
2.2	20	5544 (12)	4680 (20)	3996 (17)	3636 (13)	3600 (15)		
2.2	30	5328 (13)	4914 (12)	4608 (12)	4374 (13)	4194 (11)	4122 (12)	4158 (10)
1.8	10	6696 (13)	5652 (17)	5148 (13)				
1.8	20	6516 (13)	5436 (14)	4320 (15)	3888 (16)	3780 (14)		
1.8	30	5472 (13)	5076 (13)	4662 (13)	4356 (12)	4140 (10)	4032 (11)	4032 (11)

widths when investigating the light collection dependence on crystal length and surface finish.

Fig. 3 shows the photopeak position as a function of depth for all etched crystals averaged over the different crystal widths in order to simplify the plot. The maximum output is 6696 ± 612 , 5796 ± 432 , and 5328 ± 288 photons for 10, 20, and 30 mm long crystals, respectively, demonstrating only a weak dependence on crystal length. The loss factor is effectively independent of crystal length, implying that the photopeak position has a similar dependence on depth for different length crystals. When averaging over all crystals, we measure a 0, 18, 27, and 22% reduction at a 0, 10, 20, and 30 mm depth, respectively.

The measured pulse height resolution (i.e. the full-width at half-maximum of the 511 keV photopeak) for all crystals is also shown in Table 1 as a function of excitation depth. The photopeak resolution is measured directly from the energy spectrum after subtracting background (assumed to be a linear function as shown in Fig. 1b). Our measurements show little dependence on geometry or depth. The 10, 20, and 30 mm long crystals are measured with 13 ± 1 , 15 ± 2 , and 11 ± 1 % fwhm photopeak resolution, respectively, giving an average of 13 ± 2 % fwhm resolution for all etched crystals. These represent photopeak resolutions when the crystal is excited at a particular depth. Exciting the crystal at random depths will increase

the fwhm due to the depth-dependent light collection efficiency.

3.3. Surface finish

A previously etched crystal of each length and width is also mechanically polished and tested. In the process of polishing, the crystals are reduced in width by 0.1–0.2 mm. The results are averaged over the 4 widths (1.7, 2.0, 2.4, and 2.5 mm) and compared with the etched crystal results in Fig. 4, demonstrating that the light collection is similar for etched and polished crystals. The maximum output is 7092 ± 288 , 5544 ± 612 , and 5472 ± 504 photons for 10, 20, and 30 mm long polished crystals, respectively. (We compare this with the maximum output of 6696 ± 612 , 5796 ± 432 , and 5328 ± 288 photons for 10, 20, and 30 mm long etched crystals, respectively.) When averaging over all polished crystals, we measure a 0, 20, 28, and 27% reduction at a 0, 10, 20, and 30 mm depth, respectively. (Similarly we measure a 0, 18, 27, and 22% reduction at a 0, 10, 20, and 30 mm depth, respectively, when averaging over all etched crystals.)

The measured pulse-height resolution for all polished crystals is shown in Table 2 as a function of excitation depth. These measurements imply a minimal dependence on geometry or depth. The 10, 20, and 30 mm long crystals are measured with

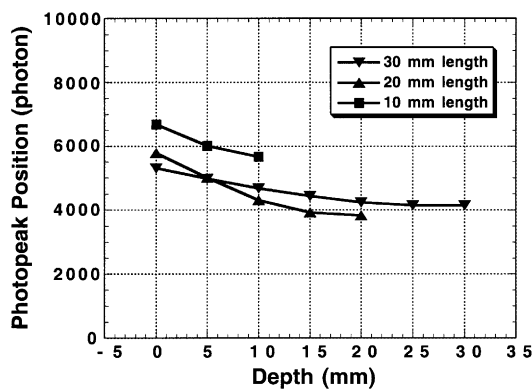


Fig. 3. The photopeak position as a function of depth (PMT end at 0 mm) for etched crystals of 3 different lengths (10, 20, and 30 mm) averaged over 4 different widths (1.8, 2.2, 2.5, and 2.6 mm) with Teflon wrapped on 5 sides.

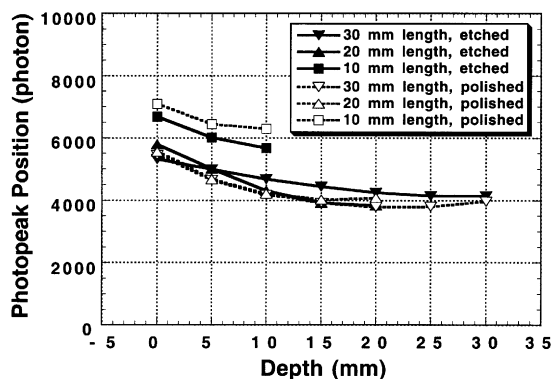


Fig. 4. The photopeak position as a function of depth (PMT end at 0 mm) for crystals of 3 different lengths (10, 20, and 30 mm) averaged over 4 different widths. Distributions are shown for both etched (solid line) and polished (dashed line) crystals with Teflon wrapped on 5 sides.

Table 2

Photopeak resolution as a function of excitation depth (PMT end at 0 mm) for polished and etched crystals of 4 different widths and 3 different lengths (10, 20, 30 mm) with Teflon wrapped on 5 sides. The photopeak resolution is listed in units of impinging photons for polished and etched (shown in parentheses) crystals

Crystal dimensions (mm)		Excitation depth (mm)						
Width	Length	0	5	10	15	20	25	30
2.6	10	15 (13)	12 (14)	12 (13)				
2.6	20	15 (17)	17 (20)	14 (19)	12 (15)	13 (13)		
2.6	30	17 (11)	17 (14)	13 (11)	14 (8)	12 (10)	14 (12)	17 (12)
2.5	10	13 (15)	14 (13)	12 (13)				
2.5	20	12 (14)	17 (15)	14 (14)	13 (14)	13 (12)		
2.5	30	14 (11)	16 (12)	13 (12)	14 (10)	12 (10)	14 (11)	13 (11)
2.2	10	14 (10)	14 (14)	11 (12)				
2.2	20	16 (12)	20 (20)	15 (17)	14 (13)	18 (15)		
2.2	30	19 (13)	16 (12)	14 (12)	15 (13)	13 (11)	14 (12)	12 (10)
1.8	10	17 (13)	11 (17)	13 (13)				
1.8	20	14 (13)	17 (14)	14 (15)	13 (16)	13 (14)		
1.8	30	13 (13)	20 (13)	15 (13)	14 (12)	15 (10)	13 (11)	15 (11)

13 ± 1 , 15 ± 1 , and $15 \pm 1\%$ fwhm resolution, respectively, giving an average of $14 \pm 1\%$ fwhm resolution for all polished crystals. (These results are comparable to the 13 ± 1 , 15 ± 2 , 11 ± 1 and $13 \pm 2\%$ fwhm resolution observed for 10, 20, 30 mm long etched crystals and their average, respectively.) Therefore, we see very similar light collection efficiency and pulse-height resolution between the etched and polished crystals.

3.4. Photodiode

We repeat these measurements using the same crystals with Teflon on 4 sides and a photodiode attached to the far (i.e. 30 mm) end, in order to test their ability to measure interaction position based on light sharing between two photodetectors. The maximum output averaged over all etched (polished) crystals with a photodiode is 4788 ± 108 (4248 ± 144) photons, representing a 19% (30%) decrease with the presence of the photodiode.

We measure a depth dependence ratio from one end of a crystal to the other. This factor is larger than that observed with Teflon on the far end due

to absorption by the photodiode (i.e. not due to additional light escaping from the reflector). This ratio is strongly dependent on etch time, varying from 7:1 from end-to-end for a 30 mm long crystal “as cut” and 3:1 for a crystal with a 5 min etch. The depth dependence ratio is again independent of width. When averaging over crystal width, we measure a depth dependence ratio of 1.6, 2.5, and 2.8 for 5 min etched crystals of length 10, 20, and 30 mm respectively (Fig. 5). Similarly we measure a ratio of 1.6, 2.0, and 2.3 for polished crystals that are 10, 20, and 30 mm long, respectively. Therefore, the end-to-end depth dependence ratio is only weakly dependent on both crystal length and these surface finishes.

4. Conclusions

We have studied the relative light collection efficiency and pulse-height resolution from narrow LSO crystals as a function of geometry and surface finish. We find that the maximum output is independent of crystal width, and weakly dependent on

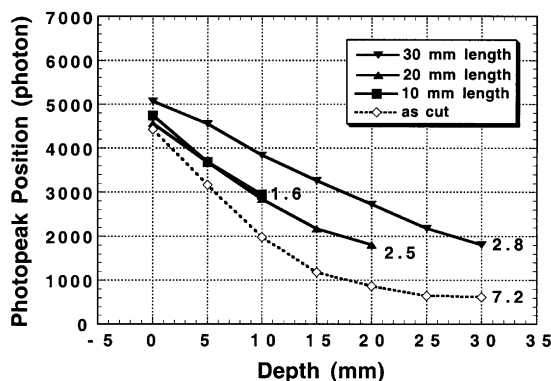


Fig. 5. The photopeak position as a function of depth for etched crystals of 3 different lengths (10, 20, and 30 mm) averaged over 4 different widths (1.8, 2.2, 2.5, and 2.6 mm), with Teflon on 4 sides and a photodiode at the far (i.e. 30 mm) end. The dashed line also shows the photopeak position for 30 mm long “as cut” (0 min etched) crystals.

both crystal length and these surface finishes. The loss factor is independent of crystal width and length, and only weakly dependent on these surface finishes. These surface treatments yield good pulse height resolution when excited at a particular depth — $13 \pm 2\%$ fwhm for etched and $14 \pm 1\%$ fwhm for polished crystals. A 5 min etch in 200°C pyrophosphoric acid gives light collection properties similar to that with a mechanical polish. We measure an end-to-end depth dependence ratio of 3:1 using crystals with a photodiode attached to the far end, as needed for our high-resolution positron emission tomograph. Since light collection depends weakly on geometry, we may choose the crystal geometry for reasons other than light collection efficiency.

Acknowledgements

This work was supported in part by the Director, Office of Science, Office of Biological and Environmental Research, Medical Science Division of the US Department of Energy under contract No. DE-AC03-76SF00098, in part by the National Institutes of Health, National Heart, Lung, and Blood Institute, National Cancer Institute, and National Institute of Neurological Disorders

and Stroke under grants No. P01-HL25840 and R01-CA67911, and in part by the Breast Cancer Fund of the State of California through the Breast Cancer Research Program of the University of California under grant No. 1RB-0068. Reference to a company or product name does not imply approval or recommendation by the University of California or the US Department of Energy to the exclusion of others that may be suitable.

References

- [1] C.L. Melcher, J.S. Schweitzer, Cerium-doped lutetium oxyorthosilicate: a fast, efficient new scintillator, *IEEE Trans. Nucl. Sci.* NS-39 (1992) 502.
- [2] J.S. Huber, W.W. Moses et al., Characterization of a 64 channel PET detector using photodiodes for crystal identification, *IEEE Trans. Nucl. Sci.* NS-44 (1997) 1197.
- [3] G. Keil, Design principles of fluorescence radiation converters, *Nucl. Instr. and Meth.* 89 (1970) 111.
- [4] W.A. Shurcliff, R.C. Jones, The Trapping of Fluorescent Light Produced within Objects of High Geometrical Symmetry, *J. Opt. Soc. Am.* 39 (11) (1949) 912.
- [5] G.F. Knoll, T.F. Knoll, light collection in scintillation detector composites for neutron detection, *IEEE Trans. Nucl. Sci.* NS-35 (1988) 872.
- [6] A. Levin, C.A. Moisan, A more physical approach to model the surface treatment of scintillation counters and its implementation into DETECT, 1996 IEEE Nuclear Science Symposium Conference Record 2, 1997, pp. 702.
- [7] S.I. Ziegler, H. Ostertag et al, Effects of scintillation light collection on the time resolution of a time-of-flight detector for annihilation quanta, *IEEE Trans. Nucl. Sci.* NS-37 (1990) 574.
- [8] C. Carrier, R. Lecomte, Effect of geometrical modifications and crystal defects on the light collection in ideal rectangular parallelepipedic BGO scintillators, *Nucl. Instr. and Meth. A* 294 (1990) 355.
- [9] A.J. Bird, T. Carter et al, The optimisation of small CsI(Tl) gamma-ray detectors, *IEEE Trans. Nucl. Sci.* NS-40 (1993) 395.
- [10] K. Murthy, C.J. Thompson et al., A study of the light output and energy resolution of small BGO crystals, *Proceedings of the 1994 IEEE Nuclear Science Symposium and Medical Imaging Conference Norfolk, VA*, pp. 1352.
- [11] S.R. Cherry, Y. Shao et al, Collection of scintillation light from small BGO crystals, *IEEE Trans. Nucl. Sci.* NS-42 (1995) 1058.
- [12] M. Casey, R. Nutt, T. Douglass, Process for fabricating tuned light guide for photoelectrons, United States patent #4750972, June 14, 1988.
- [13] K. Kurashige, Y. Kurata et al., Surface polish of GSO scintillator using chemical process, *IEEE Trans. Nucl. Sci.* NS-45 (1998) 522.